

Josephson Plasma in $\text{RuSr}_2\text{GdCu}_2\text{O}_8$

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Josephson plasma in $\text{RuSr}_2\text{GdCu}_2\text{O}_8$, $\text{Ru}_{1-x}\text{Sr}_2\text{GdCu}_{2+x}\text{O}_8$ ($x = 0.3$), and $\text{RuSr}_2\text{Eu}_{2-x}\text{Ce}_x\text{Cu}_2\text{O}_{10}$ ($x = 0.5$) compounds is investigated by the sphere resonance method. The Josephson plasma is observed in a low-frequency region (around 8.5 cm^{-1} at $T \ll T_c$) for ferromagnetic $\text{RuSr}_2\text{GdCu}_2\text{O}_8$, while it increases to 35 cm^{-1} for non-ferromagnetic $\text{Ru}_{1-x}\text{Sr}_2\text{GdCu}_{2+x}\text{O}_8$ ($x = 0.3$), which represents a large reduction in the Josephson coupling at ferromagnetic RuO_2 block layers. The temperature dependence of the plasma does not shift to zero frequency (*i.e.* $j_c = 0$) at low temperatures, indicating that there is no transition from the 0-phase to the π -phase in these compounds. The temperature dependence and the oscillator strength of the peak are different from those of other non-magnetic cuprates, and the origins of these anomalies are discussed.

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It is believed that the electronic state of high- T_c cuprates in the superconducting state is two-dimensional. Many cuprates exhibit dc and ac intrinsic Josephson effects, which reveals that the $\cdots \text{S/I/S/I/S/I/S} \cdots$ (\cdots superconductor/insulator/superconductor \cdots) -type Josephson-coupled layer model is applicable to high- T_c cuprates. In this case, the plasma of condensed carriers observed in most of the cuprates in the far infrared to microwave region is thought to be Josephson plasma. There have been many studies of this phenomenon, since it represents the interlayer phase coherence between CuO_2 layers and may be related to the mechanism of high- T_c superconductivity^{1,2,3,4,5,6,7}. Of particular importance is that it provides information about the junction parameters without the need to fabricate the mesa of an intrinsic Josephson junction; the maximum critical current j_c of the junctions is directly related to the Josephson plasma frequency ω_p by the equation $\omega_p^2 = 8\pi^2 c d j_c / \epsilon_0 \Phi_0$, where d and ϵ_0 are the width and dielectric constant of the insulating layer. It should be noted that ω_p can be deduced even from the ceramics, since the Josephson plasma can be observed by measuring the sphere resonance of powder samples as well as by conventional single crystal measurements^{2,8,9,10,11}.

Recently, much attention has been paid to $\text{RuSr}_2\text{GdCu}_2\text{O}_8$ and related materials, as the coexistence of superconductivity and ferromagnetism has been reported in these compounds^{12,13,14,15,16}. Since superconductivity and ferromagnetism are mutually exclusive, many possible superconducting order parameters have already been discussed, including the self-induced vortex state, the Fulde-Ferrell-Larkin-Ovchinnikov type, triplet superconductivity, and the π -phase^{12,13,14,15,17,18,19}. The crystal structure of $\text{RuSr}_2\text{GdCu}_2\text{O}_8$ is similar to that of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ with RuO_2 layers replacing the Cu-O chains, and the ferromagnetic order, which is observed at a Curie temperature of $T_M = 133 \text{ K}$, is attributed to the Ru moment. Although the magnetic structure of the compound has not yet been clarified, it seems that the Ru moments are anti-ferromagnetic ordered

along the c -axis and canted to the ab -plane, which makes the ab -plane weak-ferromagnetic^{20,21}. So, the system may be regarded as the $\cdots \text{S/F/S/F/S/F/S} \cdots$ (\cdots superconductor/(ferro)magnet/superconductor \cdots) -type Josephson-coupled multilayers along the c -axis. In this multilayer system, the π -phase, which has a superconducting order parameter that changes the phase by π between two adjacent superconducting layers, seems to be realized since the node at the ferromagnetic layer greatly reduces the pair breaking effects. The model calculations for this compound predict a transition from the 0-phase to the π -phase at low temperatures^{17,18,19}, and a peculiar temperature dependence of j_c ; as the temperature decreases, j_c should achieve its maximum value, decrease to zero at the transition line due to the decoupling of the junctions, and then increase again in the π -phase region¹⁹. It should be noted that this j_c temperature dependence has been realized experimentally in artificial Josephson junctions consisting of Nb and ferromagnetic $\text{Cu}_x\text{Ni}_{1-x}$ alloy as the transition from the 0-junction to the π -junction²². In the present case, it is impossible to determine j_c by I-V measurement, since no one has yet grown millimeter size single crystal of Ru-cuprates²³. The only way to determine the j_c temperature dependence seems to be to measure ω_p by measuring the sphere resonance of powder samples.

This paper reports our measurement of the far-infrared sphere resonance of $\text{RuSr}_2\text{GdCu}_2\text{O}_8$, $\text{Ru}_{1-x}\text{Sr}_2\text{GdCu}_{2+x}\text{O}_8$ ($x = 0.3$), and $\text{RuSr}_2\text{Eu}_{2-x}\text{Ce}_x\text{Cu}_2\text{O}_{10}$ ($x = 0.5$) powder samples down to 7 cm^{-1} . While all the samples exhibit the Josephson plasma resonance, the temperature dependence does not shift to zero frequency at low temperature as expected from the π transition. The peak frequency is greatly reduced in the ferromagnetic samples, indicating a considerable reduction in the Josephson coupling of the junctions. The anomalous features of the plasma are also discussed.

The ceramic samples were synthesized by the conventional solid-state reaction of oxides and carbonates un-

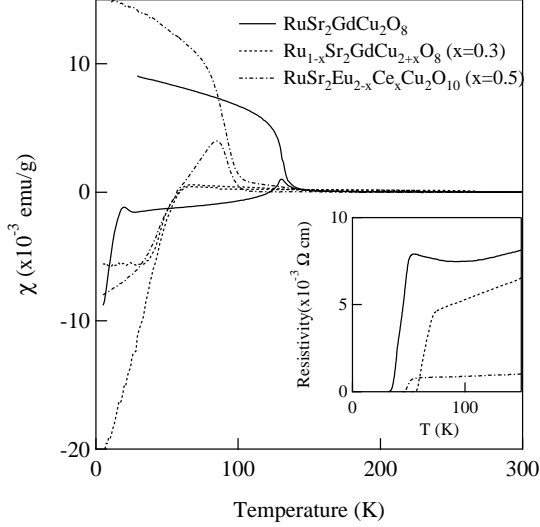


FIG. 1: Magnetic susceptibility and resistivity (inset) of $\text{RuSr}_2\text{GdCu}_2\text{O}_8$, $\text{Ru}_{1-x}\text{Sr}_2\text{GdCu}_{2+x}\text{O}_8$ ($x = 0.3$) and $\text{RuSr}_2\text{Eu}_{2-x}\text{Ce}_x\text{Cu}_2\text{O}_{10}$ ($x = 0.5$) ceramics.

der almost the same conditions described in previous reports^{12,13,14,15,24}. After several cycles of sintering and regrinding, annealing was performed at 1065°C for 150 hr at 1 atm O_2 for $\text{RuSr}_2\text{GdCu}_2\text{O}_8$, at 600°C for 50 hr at 400 atm O_2 for $\text{Ru}_{1-x}\text{Sr}_2\text{GdCu}_{2+x}\text{O}_8$ ($x = 0.3$), and at 600°C for 30 hr at 180 atm O_2 for $\text{RuSr}_2\text{Eu}_{2-x}\text{Ce}_x\text{Cu}_2\text{O}_{10}$ ($x = 0.5$). A furnace for hot isostatic pressing (HIP) was used for the high-oxygen-pressure annealing. The magnetic susceptibility and resistivity of the samples are summarized in Fig. 1. Although a magnetic transition is observed at $T_M = 133$ K for $\text{RuSr}_2\text{GdCu}_2\text{O}_8$ and around 100 K for $\text{RuSr}_2\text{Eu}_{2-x}\text{Ce}_x\text{Cu}_2\text{O}_{10}$ ($x = 0.5$), no magnetic transition is observed for $\text{Ru}_{1-x}\text{Sr}_2\text{GdCu}_{2+x}\text{O}_8$ ($x = 0.3$). The resistivity shows $T_c^{\text{onset}} = 53$ K and $T_c^{\text{zero}} = 34$ K for $\text{RuSr}_2\text{GdCu}_2\text{O}_8$, $T_c^{\text{onset}} = 64$ K and $T_c^{\text{zero}} = 56$ K for $\text{Ru}_{1-x}\text{Sr}_2\text{GdCu}_{2+x}\text{O}_8$ ($x = 0.3$), and $T_c^{\text{onset}} = 54$ K and $T_c^{\text{zero}} = 46$ K for $\text{RuSr}_2\text{Eu}_{2-x}\text{Ce}_x\text{Cu}_2\text{O}_{10}$ ($x = 0.5$). The powder X-ray diffraction indicated a single phase for $\text{RuSr}_2\text{GdCu}_2\text{O}_8$, while a slight trace of an unknown impurity phase (less than 5 %) was observed for $\text{Ru}_{1-x}\text{Sr}_2\text{GdCu}_{2+x}\text{O}_8$ ($x = 0.3$) and $\text{RuSr}_2\text{Eu}_{2-x}\text{Ce}_x\text{Cu}_2\text{O}_{10}$ ($x = 0.5$). The magnetic susceptibility, T_c , magnitude of resistivity, and x-ray results for these data agree well with previous reports^{12,13,14,15,24}. The samples were ground into fine particles about 2 μm in diameter, mixed with polyethylene powder, and then pressed into pellets about 2 mm thick. Transmission spectra of the pellets were measured down to 7 cm^{-1} using a Fourier transform interferometer combined with a Si bolometer.

Figure 2 shows the difference between the absorption coefficients of the superconducting and normal states for $\text{RuSr}_2\text{GdCu}_2\text{O}_8$ ceramics. Below T_c^{onset} , the Josephson plasma peak appears and the oscillator strength increases

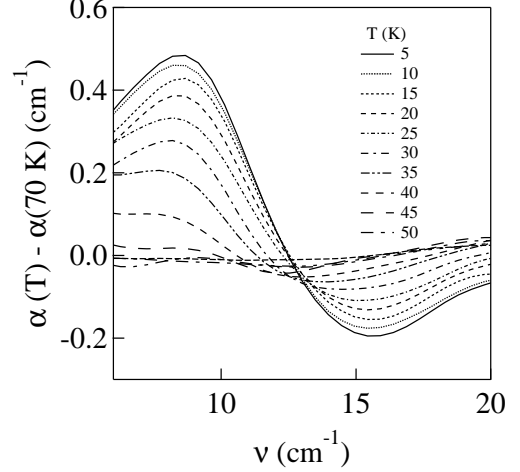


FIG. 2: Difference between α in the superconducting and normal states for $\text{RuSr}_2\text{GdCu}_2\text{O}_8$ ceramics.

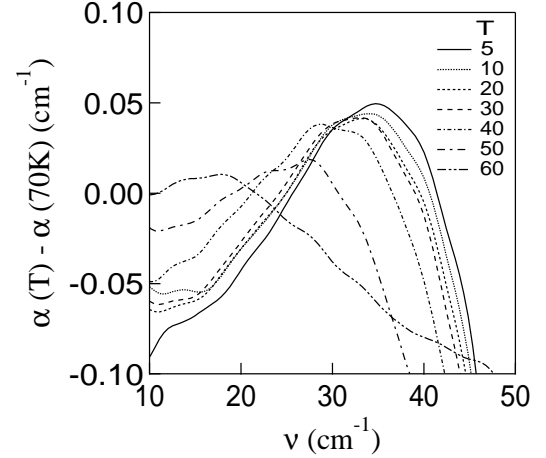


FIG. 3: Difference between α in the superconducting and normal states for $\text{Ru}_{1-x}\text{Sr}_2\text{GdCu}_{2+x}\text{O}_8$ ($x = 0.3$) ceramics.

as the temperature decreases. The peak is very broad and it is impossible to determine the peak frequency, however it becomes rather narrow below $T_c^{\text{zero}} = 34$ K. The peak is around 8.5 cm^{-1} at 5 K. The peak frequency is very low compared to that of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ plasma above 100 cm^{-1} , which has a similar crystal structure and is in a similar doping level (optimum to overdoped region)^{2,25,26,27}, and this suggests a large reduction in the Josephson coupling at the ferromagnetic RuO_2 block layers. The peak does not shift to zero frequency with decreasing temperature, which indicates that there is no 0 - π transition in this compound.

Figure 3 shows the Josephson plasma peaks of non-ferromagnetic $\text{Ru}_{1-x}\text{Sr}_2\text{GdCu}_{2+x}\text{O}_8$ ($x = 0.3$) ceramics. Compared with ferromagnetic $\text{RuSr}_2\text{GdCu}_2\text{O}_8$, the peak frequency increases greatly to 35 cm^{-1} at 5 K. This clearly indicates the large reduction in the Joseph-

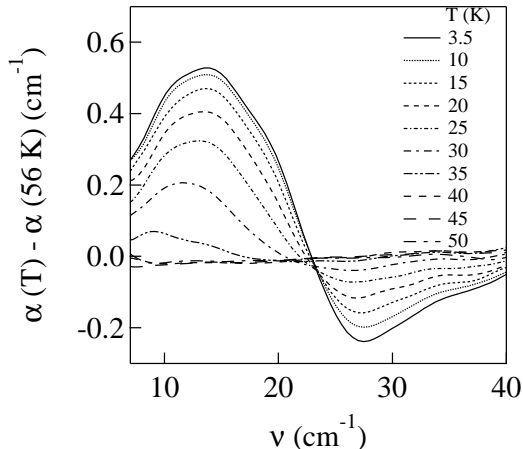


FIG. 4: Difference between α in the superconducting and normal states for $\text{RuSr}_2\text{Eu}_{2-x}\text{Ce}_x\text{Cu}_2\text{O}_{10}$ ($x = 0.5$) ceramics.

son coupling at the ferromagnetic RuO_2 block layers for $\text{RuSr}_2\text{GdCu}_2\text{O}_8$ and its absence at the non-ferromagnetic $(\text{Ru}, \text{Cu})\text{O}_2$ block layers for $\text{Ru}_{1-x}\text{Sr}_2\text{GdCu}_{2+x}\text{O}_8$ ($x = 0.3$). The peak frequencies increase as the temperature decreases, indicating the absence of a $0 - \pi$ transition in this compound. The peak oscillator strength is very weak compared with other cuprates, as discussed later.

A similar peak is also observed in $\text{RuSr}_2\text{Eu}_{2-x}\text{Ce}_x\text{Cu}_2\text{O}_{10}$ ($x = 0.5$) ceramics, as shown in Fig. 4. In this case, the peak frequency is around 13 cm^{-1} at 5 K, corresponding to the large reduction in the coupling at the magnetic RuO_2 block layers. There are two kinds of junction per unit cell in the compound; that at the RuO_2 block layers and that at the fluorite-type $(\text{Eu}, \text{Ce})\text{O}_2$ block layers, and we may expect the existence of double Josephson plasma^{5,6,7,10}, which is not seen in Fig. 4. The absence of double Josephson plasma may be explained by the degenerating plasma frequencies of both junctions, since the plasma frequency of fluorite-type block layers is around 10 cm^{-1} .³ The observed broadness of the peak also supports this explanation. Moreover, the temperature dependence of the peak does not show the $0 - \pi$ transition.

Although there is no transition to the π -phase, the plasma shows certain anomalies that are quite different from those of other non-magnetic cuprates. The first is the temperature dependence of the plasma in $\text{RuSr}_2\text{GdCu}_2\text{O}_8$; the peak frequency of the plasma is almost constant and only the peak oscillator strength increases as the temperature decreases below T_c^{zero} , which is in contrast to the monotonic ω_p increase in other cuprates. Since the peak oscillator strength is proportional to $\omega_p^2 f$, where f is the volume fraction of the superconductor in the pellet²⁸, this indicates that ω_p is almost constant while f increases as the temperature decreases. The calculation of $\cdots\text{S/F/S/F/S}\cdots$ -type multi-

layers predicts that ω_p decreases slightly at low temperatures from the monotonic increase as the exchange energy increases from zero, and the decrease becomes large as the exchange energy increases to the $0 - \pi$ transition line¹⁹. The constant behavior observed for ω_p suggests that the exchange energy, while not zero for the sample, is small and far from the $0 - \pi$ transition line.

The second anomaly is the weak oscillator strength of the plasma; the observed oscillator strength of the peak is smaller than that of other non-magnetic cuprates, especially for $\text{Ru}_{1-x}\text{Sr}_2\text{GdCu}_{2+x}\text{O}_8$ ($x = 0.3$), while we make the volume fraction f around 0.01 for all pellets. This can be explained by the very small grain size of the Ru-cuprates, which is known as granularity¹³. With the sphere resonance method, the powder (about $2\text{ }\mu\text{m}$) in the pellets is assumed to be a single crystal, and powder whose c-axis is parallel to the electric field of light causes the peak. This assumption seems not to hold for Ru-cuprates since most of the powder is formed of granules rather than a single crystal. The granular powder, which is dominated by the in-plane dielectric constant, does not form a sphere resonance peak, while the powder, which is almost composed of single domains, forms a broad peak due to the modulation of the Josephson-coupling from the in-plane dielectric constant. So, we expect the total absorption peak of granular samples to be small and broad.

The last anomaly is the strong peak broadening between T_c^{zero} and T_c^{onset} in $\text{RuSr}_2\text{GdCu}_2\text{O}_8$. One possible explanation is the granularity of the samples as discussed above. However in this case, we cannot explain the broadening change at T_c^{zero} . Another possible explanation is the melting of the self-induced vortex at T_c^{zero} , which has been suggested by other experiments^{13,25}. In this scenario, the peak is sharp in a solid phase below T_c^{zero} , and it becomes broad in a vortex liquid phase between T_c^{zero} and T_c^{onset} . Here, we also expect the peak to become sharp again in the liquid phase, which is different from the experiment.

In summary, Josephson plasma is observed at 8.5 cm^{-1} and 13 cm^{-1} for ferromagnetic $\text{RuSr}_2\text{GdCu}_2\text{O}_8$ and $\text{RuSr}_2\text{Eu}_{2-x}\text{Ce}_x\text{Cu}_2\text{O}_{10}$ ($x = 0.5$), and it increases to 35 cm^{-1} for non-ferromagnetic $\text{Ru}_{1-x}\text{Sr}_2\text{GdCu}_{2+x}\text{O}_8$ ($x = 0.3$), which indicates a large reduction in the Josephson coupling at the ferromagnetic RuO_2 block layers. Although the temperature dependence of the plasma in these compounds does not show the $0 - \pi$ phase transition, no increase in ω_p is observed for $\text{RuSr}_2\text{GdCu}_2\text{O}_8$ as the temperature decreases, which suggests a small exchange energy in the compound. The anomalous temperature dependence of the peak frequency, broadening of the peak above T_c^{zero} , and the small oscillator strength were also discussed.

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